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ART. I.—*The Relative Activity of Radium and the Uranium with which it is in Radioactive Equilibrium;**

by J. H. L. JOHNSTONE and B. B. BOLTWOOD.

Although the matter has been under consideration and discussion for a number of years, the genetic relationship between the earlier members of the uranium family of radio-elements is still a matter of considerable uncertainty. It is generally conceded that both ionium and actinium are products of the radioactive disintegration of uranium, but the exact point of origin of actinium and its immediate parentage have remained somewhat obscure and uncertain.

The work of Boltwood[1] on the relative α-ray activity of uranium minerals and the uranium which they contained demonstrated a constancy of relationship between the radioactive constituents of the older minerals and clearly indicated a close genetic relation between uranium and actinium. His determination of the activities of the more stable α-ray products relative to the activity of the associated uranium showed a simple and direct relation to exist between the products ionium, radium and polonium, but showed an abnormally low value for the ratio in the case of the actinium series, which could only be explained on the assumption that actinium originates as a branch product and belongs to what may be termed a collateral branch of the ionium-radium-polonium family.

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* The experimental results given in this paper and the general theoretical conclusions are taken from a dissertation on the "Relative Activity of Uranium and Radium" presented on April 27, 1916, by J. H. L. Johnstone in fulfillment of the requirements for the degree of Doctor of Philosophy in Yale University. The chief reason for the delay in publication was the entry of Dr. Johnstone into active military service with the Canadian forces in May, 1916. The work was carried out in the Sloane Physical Laboratory of Yale University.

[1] This Journal, 25, 269, 1908.

The values obtained by Boltwood in the course of his experiments showed that the activity of the uranium was about 2·2 times that of the radium with which it was in equilibrium, although at that time the range of the α-particle from uranium was supposed to be about 2·7 cm., which is less than that of the α-particle from radium. Since the ionizing power of an α-particle is nearly proportional to the range, and since, on the basis of the disintegration theory, an equal number of α-particles are emitted per second by each of two radioactive products in equilibrium with one another, it was necessary to assume that either the uranium atom emitted two α-particles simultaneously, which was improbable, or that two distinct α-ray changes existed in ordinary uranium. Neither of these assumptions, however, completely obviated the difficulty.

The fact that uranium actually did emit twice the number of α-particles to be expected on theoretical grounds was subsequently demonstrated by Geiger and Rutherford[2] and by Brown[3] who counted the number of α-particles emitted per second from a film of pure uranium oxide and a similar film of uraninite of known composition. Using the scintillation method, Marsden and Barratt[4] made a careful examination of the α-radiation from uranium and concluded as a result of their experiments that ordinary uranium consists of a mixture of two successive α-ray products in equilibrium with one another. Attempts to measure the separate ranges of the α-particles emitted by these two products were made by Foch[5] and Friedmann.[6] By the use of a better method of measurement, in which the Bragg ionization curves for a uranium film were compared with the corresponding curves obtained with polonium and ionium, Geiger and Nuttall calculated the ranges of the α-particles from uranium to be 2·5 cm. and 2·9 cm. (at 0°C.).

[2] Phil. Mag., 20, 691, 1910.
[3] Proc. Roy. Soc., A 84, 151, 1910.
[4] Phys. Soc. Proc., A 23, 367, 1911.
[5] Le Radium, 8, 101, 1911.
[6] Wien. Ber., 120, 1361, 1911.

Numerous unsuccessful attempts have been made to reduce the specific α-ray activity of uranium. In one experiment conducted by the authors about two kilograms of pure uranium nitrate were subjected to fractional crystallization and a least soluble "head" fraction weighing about twenty grams was obtained after about forty operations. The specific α-ray activity of the uranium in this material did not vary by as much as one per cent from the specific activity of the uranium in the original nitrate. This shows that the two components are so closely allied chemically as to be inseparable, a conclusion which is supported by all the other known facts at our disposal.

We may outline, therefore, the progressive disintegration of the uranium atom, considering for the present only the products emitting α-rays, as taking place in the following manner: the parent element, uranium I, produces the product uranium II. This in turn produces ionium, which disintegrates to form radium, followed successively by radium emanation, radium A, radium C and polonium. When these are all present in equilibrium proportions, as is the case in a non-emanating, old, radioactive mineral, then certain comparatively simple relations will exist between the α-ray activities of the different constituents. It has been shown by Geiger[7] that the ionizing power of an α-particle is proportional to the two-thirds power of its range. The ionization produced by equal numbers of α-particles emitted by two different radio-elements will therefore be proportional to the two-thirds power of the ranges of these particles. In a series of successive products in equilibrium, each product emits the same number of α-particles in unit time. The relative ionization (and therefore the relative activities) due to each of these products should therefore be proportional to the two-thirds power of the range of the respective α-particles.

This relation has been shown[8] to hold quite closely in the case of radio-thorium and its α-ray products, and also in the actinium and the radium series of products. The chief object of the work described in the present paper was to apply the same methods to the case of the uranium-radium series with the expectation that the results would throw some light on the obscure relations of the earlier members of the series.

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[7] Proc. Roy. Soc., A 83, 505, 1910.

[8] McCoy and Viol, Phil. Mag., 25, 333, 1913; McCoy and Leman, Phys. Rev., 4, 409, 1914; ibid. 6, 185, 1915.

The Radioactive Measurements.

The determinations of the radioactivity of the different solids examined were carried out in an electroscope which has already been described.[9] In the present experiments a telemicroscope was used for observing the position of the gold-leaf. The natural leak of the instrument was small and over a period of about six months varied from 0·4 to 0·7 scale divisions per minute. Before and after each series of measurements the sensibility of the electroscope was determined by measurements of the activity of a standard reference film of pure uranoso-uranic oxide. This film was carefully preserved throughout the entire period of the measurements here recorded and all the results given in this paper are given in terms of this film as the standard.

The method of preparing the radioactive materials for measurement was essentially the same, with certain modifications, as that described by Boltwood. The material to be examined was ground as finely as possible in the form of a thin paste with pure ethyl alcohol in a small agate mortar. A sheet of aluminium 7·5 × 9 cm. and 0·01 cm. thick was first carefully cleaned with liquid soap and distilled water and was then placed in a drying oven at 65°C. for 15 minutes. It was placed in a desiccator over sulphuric acid for half an hour, and then weighed on a sensitive chemical balance. The paste of material and alcohol was then thinly spread on the surface of the aluminium with a small camel's-hair brush.[10] The coated plate was placed in the oven, cooled in the desiccator and weighed as before. The weight of the films could be determined in this manner with an accuracy of one per cent. The solid material adhered quite strongly to the plate and showed no tendency to fall off even when the plate was inverted.

The measurements of radium emanation were made with a gold-leaf electroscope having an air-tight ionization chamber with a capacity of about three liters. The separation and collection of the radium emanation, its transfer to the electroscope and the measurement of its radioactivity were carried out according to methods which have already been described.[11]

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[9] Boltwood, this Journal, 25, 272, 1908.

[10] The brushes used were carefully cleaned in advance by long immersion in alcohol and subsequent washing in fresh quantities of the same liquid.

Ratio of the Activity of a Uranium Mineral to the Activity of the Contained Uranium.

The relation of the activity of the parent element, uranium, associated with equilibrium amounts of all of its disintegration products, to the activity of the parent element alone, is a fundamental quantity of great importance to any consideration of the relations existing between the individual products themselves. The actual progenitor of the series is uranium I, but this can not be isolated from its invariable associate and isotopic product, uranium II. The combined effect of these two elements when mixed in equilibrium proportions can be determined, however, and this can be compared with the activity of a similar mixture containing all the other disintegration products in equilibrium proportions.

Such a mixture is furnished by a pure, primary, unaltered uranite. A mineral containing a low proportion of thorium is preferable since a correction must be made for the activity of the thorium products present. A specimen of uraninite from Spruce Pine in the possession of the authors was considered to fulfill all the necessary requirements. It consisted of essentially unaltered material selected with much care from a considerably larger quantity. It contained less than 0·2% of silica and residue insoluble in dilute nitric acid. A determination of the uranium content was made by one of the authors and by Ledoux and Co. of New York City.[12] The mineral contained 1·9% of thorium oxide.

In determining the activity of uranium a very pure specimen of uranoso-uranic oxide was used. This had been prepared from a specimen of especially pure uranium nitrate obtained by fractional recrystallization of a much larger quantity (see p. 3). The oxide was made from the nitrate with all the precautions which have been mentioned in an earlier paper.[13] It is very important to note that this oxide was used as a standard in the analytical determination of uranium (both volumetric and gravimetric) in the mineral, so that the analytical determinations and radioactive measurements are in direct accord with one another, although it was assumed for purposes of calculation that the uranoso-uranic oxide contained 84·8% of uranium.

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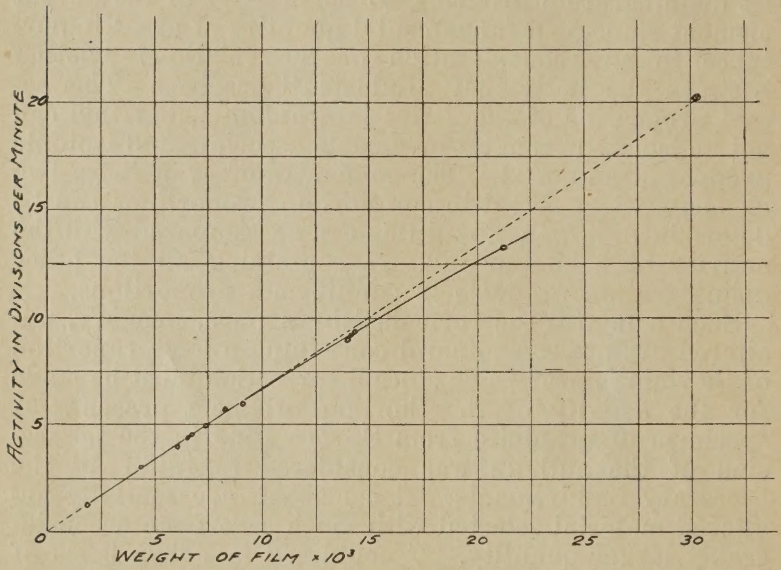
[11] Boltwood, this Journal, 18, 378, 1904; Phil. Mag., 599, 1905.

[12] The authors wish to express here their obligation to Ledoux & Co. for this favor and to state their appreciation of the value of this carefully conducted analysis.

It has been found by McCoy[14] that considerable absorption of the radiation takes place in the film itself when its thickness is appreciable. In earlier work by one of us[15] it was shown that by the use of thin films any necessity for an absorption correction could be avoided. To further demonstrate this fact a series of films of uranoso-uranic oxide weighing from 0·0019 g. to 0·04 g. were prepared and their activities were measured. The results are given in table I and are shown graphically in Fig. 1. In films weighing not more

Fig. 1.



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[13] Boltwood, this Journal, 25, 278, 1908.
[14] Jour. Am. Chem. Soc., 27, 391, 1905; Phys. Rev., 1, 393, 1913.
[15] Boltwood, this Journal, 25, 176, 1908.

than ten milligrams the absorption of the α-radiation was negligible. In all cases where values of importance were to be derived the weight of the films used was less than this maximum, so that no correction has to be made in the results for absorption of the radiations.

The average of the first eight values in the fourth column of table I is 666, which denotes that the average activity of the eight lighter films was 666 divisions per minute per gram of uranium oxide. This corresponds to an activity of 785 (viz. 666/0·848) divisions per minute per gram of uranium.

Table I.

Film Number	Weight of oxide in grams	Activity Div./Min.	Activity Weight
20	·00187	1·24	663
19	·00432	2·91	673
15	·00566	3·77	666
18	·00653	4·37	669
21	·00740	4·95	668
22	·00832	5·56	662
14	·00898	5·95	662
23	·00653	4·38	670
25A	·01405	8·89	633
8	·01380	8·94	648
25B	·01440	9·30	645
26	·02150	13·45	624
24	·04350	25·96	596

The specific activity of the uraninite was determined by the measurement of four films weighing from approximately 2 to 6 milligrams. The results are given in table II.

Table II.

Film Number	Weight gram	Activity in divisions per minute per gram
27	0·00387	2627
28	0·00240	2655
29	0·00588	2627
30	0·00170	2600

These results give the mean value for the specific activity of the uraninite as 2624 divisions per minute per gram.

As already stated the mineral contained 0·019 gram of thorium oxide (and other thorium products in equilibrium with this) per gram of mineral. The above value for the specific activity of the mineral includes the increment due to the thorium products which must be eliminated. To accomplish this measurements were made of a series of films prepared from a specimen of thorite containing 52 per cent of thorium oxide and 0·37 per cent of uranium. The results are tabulated in table III.

Table III.

Film Number	Weight gram	Activity per gram in divisions per minute
39	0·0056	511
40	0·0076	515
42	0·00776	514
		Av. 513

The activity of the uranium and its radioactive products contained in the thorite will closely equal

$$\frac{2624}{0\cdot73} \times 0\cdot0037 = 13\cdot3 \text{ divisions per min.}$$

Correcting the specific activity of the thorite by this number and dividing the remainder by 0·52 (the weight of thorium oxide per gram) we obtain 960 divisions per minute as the activity of one gram of thorium in equilibrium with its products. We may now correct the value found for the activity of the uraninite by an amount equal to the activity of the thorium components (960 × 0·019) and obtain the value 2606 divisions per minute per gram of mineral for the uranium series of products which it contains and a value of 3570 divisions per minute per gram of uranium present (2606/·73).

The uraninite, however, contained less than the full equilibrium amounts of the uranium-radium products because of the fact that in the finely divided form in which it was used it spontaneously lost a small proportion of its radium emanation. The loss of this and the absence of the proportionate amounts of radium A and radium C would cause a deficiency which must be corrected for.

The relative proportion of radium emanation lost by the films of uraninite was determined by the method described by McCoy and Leman.[16] It was found to be 9·1 per cent. In applying the correction, the value for the ratio of the activity of the radium products (radium emanation, radium A and radium C) to the activity of the radium with which they are in equilibrium, as found by McCoy and Leman, namely 4·11, was made use of, as was also the ratio of the activity of radium to the activity of the uranium with which it is in equilibrium (0·49) which was derived in the course of the present investigation (see p. 12).

The correction has the following form,

$$3570 + (785 \times 4{\cdot}11 \times 0{\cdot}49 \times 0{\cdot}091) = 3715,$$

which gives an activity of 3715 divisions per minute per gram of uranium as the activity of the mineral due to uranium and its products in a complete state of equilibrium.

From this final result we are able to calculate the value sought, namely, the ratio of the activity of the uranium with its equilibrium amounts of disintegration products to the activity of the uranium (uranium I + uranium II) alone.

This is 3715/785 = 4·73, which is in good agreement with the value 4·69 found earlier by Boltwood.

Ratio of the Activity of Radium to the Activity of the Uranium with which it is Associated.

If the transformation of the atoms of uranium I into atoms of uranium II takes place directly without the production of any side products, and the transformations uranium II—ionium—radium proceed in the same direct manner, then the relative activities of the three members—uranium I, uranium II, radium—should be proportional to the two-thirds power of the ranges of the α-particles emitted by the respective elements, namely, the activities should be proportional to

$$(2{\cdot}37)^{\frac{2}{3}},\ (2{\cdot}75)^{\frac{2}{3}},\ (3{\cdot}13)^{\frac{2}{3}}$$

where the numbers in parentheses are the ranges at 0°C. of the α-particles from uranium I, uranium II and radium, respectively. Any departure from this proportionality will indicate an irregularity in the mode of

[16] Phys. Rev., 6, 185, 1915.

transformation and may serve to suggest the nature of the changes which are actually taking place. If the transformations are all simple the activity of the uranium (consisting of an equilibrium mixture of uranium I and uranium II) should be to the activity of the radium in the proportion

$$(1{\cdot}78 + 1{\cdot}96) : 2{\cdot}14 = 1{\cdot}00 : 0{\cdot}57.$$

An experimental determination of this ratio was carried out in the following manner:

A quantity of radium was separated from Colorado carnotite and was carefully freed from other radioactive substances which can be separated by chemical operations. Since carnotite is free from thorium the specimen of radium obtained did not contain any appreciable amounts of mesothorium or other products. A solution of this radium in dilute hydrochloric acid was then prepared and its approximate strength in radium was determined by the emanation method. Using this first solution as a basis, two other solutions (denoted hereafter as solutions B and C) were prepared which contained about 0·025 g. of barium chloride and $2{\cdot}4 \times 10^{-8}$ g. of radium in 10 cc. of solution. The quantities of radium were so chosen that the radium films ultimately obtained would have activities of the same order of magnitude as the activities of the uranium films with which they were to be compared.

An accurate determination was then made of the emanation in equilibrium with the radium in 10 cc. of the radium-barium solutions. The results were recorded in terms of the leak produced in the emanation electroscope in divisions per minute.

The results were

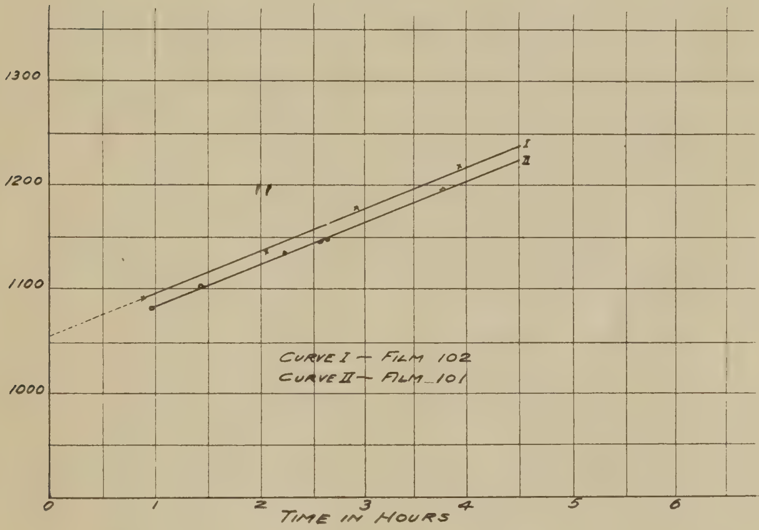
For 10 cc. of Solution B...........	81·7 div. per min.
For 10 cc. of Solution C...........	71·0 div. per min.

The radium emanation in equilibrium with the radium contained in one gram of the uraninite was also determined and was found to correspond to 853·5 divisions per minute in the same electroscope. Since the uraninite contained 73·0% of uranium, the radium in equilibrium with one gram of uranium in the mineral was equivalent to a leak of 1170 divisions per minute. The quantity of radium in 10 cc. of solution B, therefore, was the same as that in equilibrium with 0·0698 g. of uranium, and in 10 cc. of solution C with 0·0607 g. of uranium.

The α-ray activity of the radium itself was determined in the following manner:

Portions of solutions B and C 10 cc. in volume were taken, and the barium and radium present were precipitated as sulphate under standard analytical conditions. The precipitates were removed and ignited as promptly as possible and the time at which the ignition was carried out was noted. The weight of the precipitate was determined and the material was quickly ground to a fine powder with ethyl alcohol. Films of this material were then prepared in the manner already described and the activity of these films was measured in the α-ray electroscope. Measurements of the activity were continued over a period of 6 to 7 hours and the increase with the time (due to the growth of the emanation and other active products) was noted at definite intervals.

Fig. 2.



Preparatory to the precipitation of the sulphates the radium-barium solutions were kept at a temperature slightly below the boiling point for a period of four hours in order to remove any emanation *as formed* and to permit the other products (radium A, radium C) to completely disintegrate. The activity of the material forming the films was therefore due to radium alone at the time of ignition, and this activity could be easily determined from the data available. The variation of the activity with the time for two typical films is shown in figure 2, where the zero time is taken as the moment of ignition. By a simple extrapolation of the curve the initial activity could be obtained with accuracy.

The calculation of the ratio of the activity of the radium to the activity of the uranium was made by the use of the following equation, where

x = the required ratio.
Em_1 = the activity of the equilibrium amount of emanation from one gram of uraninite in div. per min. (in the emanation electroscope).
Em_2 = the activity of the equilibrium amount of emanation from 10 cc. of radium-barium solution in div. per min. (in the emanation electroscope).
U = the activity of one gram of uranium oxide in div. per min. (in the α-ray electroscope).
R = the initial activity of one gram of radium-barium sulphate (in the α-ray electroscope).
w = the weight of sulphate precipitated from 10 cc. of the radium-barium solution.

The value of x is given by,

$$x = \frac{\mathrm{W} \times \mathrm{Em}_1 \times \mathrm{R} \times 0{\cdot}848}{0{\cdot}73 \times \mathrm{Em}_2 \times \mathrm{U}}.$$

The advantage of this method of calculation lies in the fact that the question of either radium or uranium standards is not involved in the final value.

The results obtained from the measurement of the radium films are given in table IV.

Table IV.

Solution	Film Number	Film Weight	U	R	W	Em_1	Em_2	X
B	101	0·00926	710[a]	1040	0·0278	853	81·8	0·493
B	102	0·00711	710[a]	1050	0·0278	853	81·7	0·497
B	103	0·00819	710[a]	1030	0·0277	853	81·7	0·487
B	104	0·00667	710[a]	1025	0·0274	853	81·8	0·480
C	110	0·00548	710[a]	1025	0·0237	853	71·0	0·478
C	113	0·00300	710[a]	1050	0·0237	853	70·0	0·495

Mean value of $x = 0{\cdot}488$

[a] The value here given is based on a different sensibility of the electroscope from that which it possessed in the case of the values given in table 1.

The value obtained by Boltwood[17] for this ratio (0·45) is somewhat lower than that given above. When the experimental conditions are taken into consideration, however, the agreement is as good as might be expected. A determination has also been made by Meyer and Paneth[18] who compared the radiation from a known quantity of radium with the ionization produced by the α-particles from one gram of uranium. They obtained a value of 0·57 for the ratio. Aside from other objections to their method, the manner in which they obtained the uranium salt used as a standard is open to the most serious criticism. Comparatively crude uranyl nitrate was subjected to purely chemical methods of purification, methods which are generally recognized as unsuitable for obtaining a pure uranium product. Very little, if any, weight can therefore be attached to their determination of the value of the ratio.

Discussion of Results.

As already pointed out, if the entire series of transformations from uranium I, through uranium II and ionium, to radium is a simple and direct one, the value to be expected for the uranium-radium ratio is approximately 0·57. The value found in this investigation is 0·49, which is lower by an amount far in excess of the probable experimental error. The result suggests that the number of radium atoms which disintegrate with the emission of α-particles in the unit time is less than the number of atoms of uranium I or uranium II which disintegrate in the same period. This indicates either (*a*) that a series of branch products is split off from the main series before the radium atom is produced, or (*b*) that radium itself disintegrates in a complex manner, a larger proportion (but not all) of the atoms being transformed with the emission of α-particles. Unless the accepted values for the ranges of the α-particles from uranium are greatly in error (which appears to be rather improbable) the progress of transformation from uranium I to (and including) radium is at some point irregular and is accompanied by the production of a collateral series. This conclusion is supported by the occurrence of actinium and its products in association

[17] This Journal, 25, 269, 1908.
[18] Wien. Ber., 121, Abt. IIa, 1912.

with radium in uranium minerals, and by the impossibility of tracing the origin of actinium to any point in the series subsequent to radium.

We will now proceed to a consideration of the two alternatives, (*a*) and (*b*) mentioned in the preceding paragraph. As a preliminary to this the relative activities of the other members of the main-line series can be calculated from the ratio experimentally determined for the uranium and radium. Geiger's equation is employed

$$I = Nk\,R^{\frac{2}{3}}$$

and the results obtained are given in table V. The calculations are made on the assumption that the simplest (1:1) relation exists between all the products following and including ionium.

Table V.

Element	Range of α-particle in cm. (at 0°C.)	Relative Activity Calc.	Found
Uranium I	2·37		
Uranium II	2·75	1·00	1·00
Ionium	2·95	0·46	
Radium	3·13	0·49	0·49
Emanation	3·94	0·57	
Radium A	4·50	0·62	
Radium C	6·57	0·80	
Radium F	3·64	0·53	

Sum = 4·47

Relative activity of actinium and actinium products as determined experimentally by Boltwood.[19] = 0·28

Total = 4·75

Total activity as determined (p. 9) .... 4·73

The *very* close agreement between the total activities as calculated and as found is purely accidental and merely indicates that the value given for the activity of the actinium products by difference (4·73–4·47 = 0·26) is practically the same as that suggested by Boltwood as a result of his experiments.

The ranges of the α-particles from uranium, radium and each of the actinium products and also the value of the two-thirds power of the range are given in table VI.

[19] This Journal, 25, 297, 1908.

Table VI.

Element	Range at 20°	$R^2/_3$
Uranium I	$R_1 = 2{\cdot}54$	1·87
Uranium II	$R_2 = 2{\cdot}95$	2·05
Radium	$R_3 = 3{\cdot}36$	2·24
Radioactinium	$R_4 = 4{\cdot}29$	2·64
Actinium X	$R_5 = 4{\cdot}34$	2·66
Actinium emanation	$R_6 = 5{\cdot}66$	3·18
Actinium A	$R_7 = 6{\cdot}37$	3·44
Actinium C	$R_8 = 5{\cdot}24$	3·01

Applying the data available we may calculate what proportion of the total number of atoms of uranium II would have to be assumed to disintegrate in a mode leading to the production of actinium in order that the ratio of the activity of the actinium products to the activity of the radium would have the value 0·28/0·49 indicated in table V. There are five actinium products emitting α-rays as compared with a single α-ray change in the case of radium, and if equal numbers of atoms of each of the elements were disintegrating in unit time the ratio of the activities would be[20]

$$(2{\cdot}64 + 2{\cdot}66 + 3{\cdot}18 + 3{\cdot}44 + 3{\cdot}01) : 2{\cdot}24 = 6{\cdot}6 : 1.$$

The observed ratio is, however, 0·28 : 0·49. If 100 atoms of uranium II are assumed to disintegrate in unit time of which x disintegrate to form actinium, we have the relation

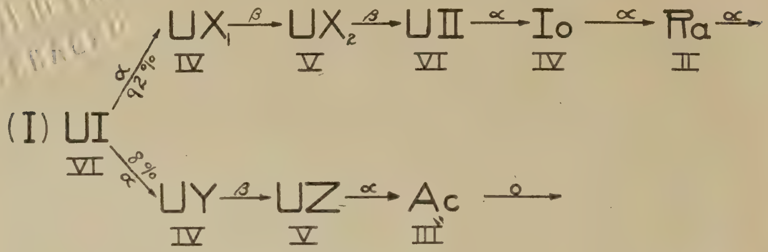
$$\frac{14{\cdot}9\,x}{(100 - x)\,2{\cdot}24} = \frac{0{\cdot}28}{0{\cdot}49}$$

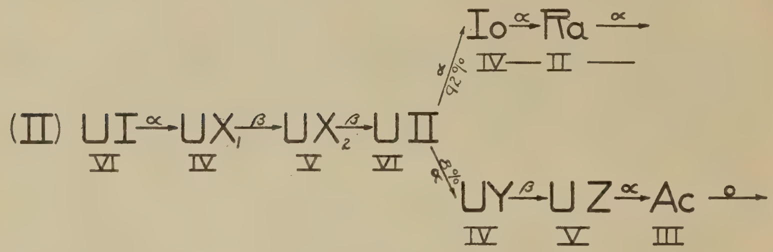
which gives a value for x of approximately 8. So that, if, out of every one hundred atoms of uranium II disintegrating, a total of eight atoms changed into actinium and the remaining 92 changed into ionium (and ultimately radium) the observed relations would exist between the activity of the radium and the activity of the actinium products in a mineral.

Based on considerations of this character a number of attempts have been made to devise a scheme of transformation which will satisfactorily indicate the successive changes undergone by the uranium atoms. The most plausible of these have been proposed by Soddy and Cranston[21] and are given on the following page.

[20] Table VI.

[21] Proc. Roy. Soc., A 94, 384, 1918.





It is not proposed to discuss these schemes in detail or to consider at any length certain minor points involved to which exception might be taken. It is, however, difficult to understand just what is implied by the dual transformation with the expulsion of α-particles in both cases which is suggested for U I in the first scheme and for U II in the second. It would appear that the loss of an α-particle in each case should lead to the production of one and the same kind of matter, namely, to a single product and not to two different products to be designated as U X_1 and U Y in one example and as Io and U Y in the other. The main point under consideration is, however, whether either of these schemes give us a clue to the explanation of the relative activities of uranium and radium as they have been found in our experiments, and it may be stated that they do not, since the first scheme would suggest a ratio of 1/·55 for the relative activities of the uranium and the radium while the second scheme would imply a ratio of 1/·53 for the same quantities.

In order that Scheme I might satisfactorily apply to the ratio as found by experiment it would be necessary to assume that about 26 out of every 100 atoms of U I were transformed in the mode leading to the production of actinium. This in turn is contradicted by the relative activity of the actinium products in an equilibrium mixture.

In order to fulfill the conditions involved in Scheme II it would be necessary for 14 out of every 100 atoms of uranium to be transformed in the mode leading to the production of actinium. Nor is there any apparent advantage gained by assuming that the transformation of either U I or U II into the first member of the side series is accompanied by the expulsion of a β-particle instead of an α-particle.

It might, however, be assumed that the branching of the series takes place at some other point, as at radium, for example, and that 86 per cent of the radium atoms disintegrate with the emission of α-rays to form the emanation, etc., while 14 per cent disintegrate (emitting β-rays) to form actinium. Direct evidence of the emission of β-rays by a specimen of radium has been obtained by Hahn and Meitner.[22] Under these conditions the α-ray activity of ionium would be proportional to the uranium radiation and would equal 0·53. The activity of the actinium series would equal 0·56 and the activity of the radium + emanation + radium A, C and F would be 3·01 (the sum of the values given in table V). The sum of all of these together with uranium is 5·10 for the total activity of the uranium series (as in uraninite). There are, however, serious objections to the assumption that the side branch arises at radium, aside from the fact that the values mentioned are widely different from those found in Boltwood's experiments and the value found in the present experiments for the total activity of the uranium products.

The most significant objection is presented by the agreement of the value found for the disintegration constant or radium by Rutherford and Geiger[23] and the value of this constant found by Miss Gleditsch.[24] The Rutherford and Geiger estimate was based on the number of α-particles emitted per second by the radium C in equilibrium with one gram of radium. If only eighty-six out of every hundred of the radium atoms disintegrate to form radium C, then this estimate would be 14 per cent too low. The method employed by Miss

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[22] Physik. Zeitschr., 10, 741, 1909.

[23] Rutherford, Phil. Mag., 28, 326, 1914.

[24] This Journal, 41, 112, 1916.

Gleditsch depended on the production of radium from the ionium in equilibrium with a known amount of radium and was measured in terms of the fraction of the total equilibrium amount which was produced in a known period of time. This would have given the true value for the disintegration constant irrespective of the mode of disintegration. These two methods would therefore have given different and not similar values if the collateral series had originated at radium.

Among other objections may be mentioned the experiment made by Soddy[25] who examined a specimen of radium salt containing 13·2 mgs. of radium which had been sealed for a period of 10 years. No evidence of the presence of actinium was obtained. Paneth and Fajans[26] examined a specimen containing 180 mgs. of radium which had been sealed for six years, but were unable to detect the presence of any actinium products. We may therefore dismiss the possibility of the side chain splitting off at radium as highly improbable in the light of our present knowledge.

The possibility that the collateral series originates at ionium may also be considered. The fact that the experimental evidence is all opposed to the emission of a β-radiation by ionium is in itself a decided objection to this view. Moreover, it would require (uranium taken as unity) an activity of 0·56 for the actinium products, an activity of 0·46 for ionium, and an activity of 3·01 for radium and its products, with a total activity of 5·02. Paneth and Fajans[27] have directly attacked this problem by seeking for the presence of actinium in a strong preparation of ionium-thorium which had been undisturbed for four years. They were unable to discover the presence of any actinium products. Lacking any support, therefore, the supposition that the collateral series arises at ionium is untenable at present.

These circumstances compel a return to a consideration of the earlier members of the series, to U I and U II, in the hope of being able to find there an explanation of the conditions indicated by our experiments. At first sight it might seem that the conditions would be satisfied

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[25] Nature, 91, 634, 1913.

[26] Wien. Ber., 123, IIa, 1627, 1914.

[27] Wien. Ber., 123, IIa, 1627, 1914.

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by assuming that what we now call uranium consists of three radio-elements, a parent element and two isotopic products in equilibrium, all emitting α-rays. But if these are present in relative amounts of the same approximate order of magnitude (*i. e.,* 100, 92, 92, etc.) then the α-rays emitted by at least one of them would have to be of exceedingly short range and small ionizing power and the rate of change of this substance would be excessively slow. It is not impossible, but it does not seem probable, that ordinary uranium may consist of what we know as U I and U II, both radio-elements in the main line of descent, and a third isotope which is a product in the collateral actinium series. But the difficulties here are not inconsiderable aside from the fact that the existence of such an isotope is somewhat difficult to imagine. If present in amounts proportional to the actinium this product would have to emit comparatively long range (7·2 cm.) α-particles and would therefore have a very short life period. Such a conclusion does not seem at all probable in the light of our present knowledge.

It is not impossible that the values accepted for the ranges of the α-particles from uranium are considerably in error and that this is the reason for the lack of agreement between theory and experiment. But until some more definite data have been obtained there seems to be little justification for abstruse speculation on the genetic relationship in the earlier stages of the uranium series.

Summary.

The relation of the activity of radium to the activity of the uranium with which it is in radioactive equilibrium has been redetermined. The results obtained indicate that if the activity of uranium is taken as unity the activity of the radium is equal to approximately 0·49.

The total activity of uranium mixed with equilibrium quantities of its disintegration products has been compared with the activity of the uranium alone, and the former has been found to be 4·73 times the latter.

A critical examination has been made of the various theories which have been proposed to explain the genesis of radium and actinium from uranium. None of these theories appears to satisfy the necessary requirements.

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